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Foreign Technology Division
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by

R. S. Saringyulyan, K. A. Kostanyan
and Ye. A. Yerznkyan

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13. ABSTRACT

Exptl. data indicate the absence of a rectilinear relationship between log sigma (elec. cond.) and log eta (viscosity). This correlation holds good in the high temp. regions. The values of n and C (consts. in Littleton equation log eta + n log sigma = C) for different glasses are presented. Glasses become inhomogeneous in anomalous temp. regions. at firing temp. regions the correlation between sigma and eta develop an exponential function.
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О о	*О о*	O, o	Ю ю	*Ю ю*	Yu, yu
П п	*П п*	P, p	Я я	*Я я*	Ya, ya

* <u>ye</u> initially, after vowels, and after ъ, ь; <u>e</u> elsewhere. When written as ё in Russian, transliterate as yё or ё. The use of diacritical marks is preferred, but such marks may be omitted when expediency dictates.

VISCOSITY AND ELECTRICAL CONDUCTIVITY OF MOLTEN GLASSES OVER A WIDE TEMPERATURE RANGE

R. S. Saringyulyan, K. A. Kosanyan
and Ye. A. Yerznkyan

The relationship between viscosity (η) and electrical conductivity (κ) of molten glasses and salts is usually given by the Littleton-Yevstrop'yev equation [1]

$$\kappa^n \eta = C, \tag{1}$$

where n and C are constants.

Data have recently been published concerning molten glasses whose viscosity does not exceed 10^5 poise [1, 2]. Frenkel, Yevstrop'yev, and other researchers consider relationship (1) to be the formal corollary of identical temperature dependences of viscosity and electrical conductivity [1, 3];

$$\eta = A_\eta e^{\frac{E_\eta}{kT}}$$

and (2)

$$\kappa = A_\kappa e^{-\frac{E_\kappa}{kT}}.$$

There is another point of view which has been discussed in the literature, one which analyses equation (1) as a corollary of the great physical connection between viscosity and electrical conductivity [2, 4].

Establishment of the actual functional dependence between viscosity and electrical conductivity requires a review not of the motten state separately, but of the entire temperature range encompassing a change in viscosity from 10^2 to 10^{14} poise in which equations (2) are no longer valid. The temperature dependences of viscosity and electrical conductivity within the indicated interval are described by more complex equations containing three or more constants. Here the equations for viscosity and electrical conductivity are also similiar in form. It is a known fact that, for example, electrical conductivity in the temperature interval encompassing the molten and highly-viscous states (the temperature range of annealing) satisfies the equations [5]

$$\lg \varkappa = A' - \frac{B'}{T - T}, \tag{3}$$

$$\lg \varkappa = A'' - \frac{B'' e^{\frac{C}{T}}}{T}, \tag{4}$$

$$\lg \varkappa = A''' - \frac{B'''}{T^n}. \tag{5}$$

which in form resemble the corresponding equations of Tamman-Vulcher, Waterton, and Okhotin. If we start with these derived equations, then the conditions for satisfying relationship (1) become more complicated. When viscosity and electrical conductivity are subject to an exponential dependence the conditions for fulfilling relationship (1) is [3]

$$E_\eta = nE_\varkappa, \tag{6}$$

while over a wide temperature range, beginning with equation (5) and the Okhotin equation

$$\lg \eta = A'''_{\eta} + \frac{B'''_{\eta}}{T^{m}}, \qquad (7)$$

the conditions for fulfilling relationship (1) will be

$$m = m' \text{ and } B'''_{\eta} = nB'''. \qquad (8)$$

If, however, we begin with equations (3) and (4) and the corresponding viscosity equations, in these cases two equalities must also be satisfied.

At present there are no calculation methods for determining the constants in equations (3-5) and the analogous equations of viscosity: this situation prevents us from verifying the feasibility of relationship (1). Exisiting literature does not indicate the methods for calculating viscosity and electrical conductivity of glasses over a wide temperature range. Existing data concern only a limited temperature range. Therefore, the sole means for verifying the feasibility of relationship (1) over a wide temperature range is experiment.

The purpose of this work is an investigation of the temperature dependences of viscosity and electrical conductivity of glasses and also the forms of the dependence between these properties in the viscosity range of 10^2 to 10^{14} poise. For the purpose of experiment, 15 glasses of different composition (among them 7 of industrial composition) were selected, several of which are shown in Table 1.

The viscosity of the glasses in the annealing temperature range was determined by vertical elongation [6], while in the molten state – by the restrained-sphere method [7]. The electrical conductivity of all the glasses over the wide temperature range

(500-1400°) was measured using the method described in work [8], modified somewhat. The obtained experimental data supported the belief that over a wide temperture range the electrical conductivity is well defined by equations (3-5) while viscosity is well defined by the Tamman-Vulcher, Waterton, Yevstrop'yev, [6], and Okhotin equations. We propose in addition the following equations for viscosity:

$$\lg \eta = a + bT + C/T^2; \quad (9)$$

$$\lg \eta = \alpha + \frac{\beta}{T} + \gamma T. \quad (10)$$

Table 1. Composition of glasses, wt %.

Glasses	Oxides							
	SiO_2	B_2O_3	Al_2O_3	CaO	MgO	BaO	K_2O	Na_2O
№31	65	·	...	10			25	
№21	65	..	—			10	25	
Lilly	70		...	9		·	—	21
E	51.8	10.8	14.5	15.9	4.0		·	·
№3	71.0	0.1	3.0	6.9	2.9		0.9	15.2
№6	68.0	0.2	5.9	6.9	3.0	·	1.9	14.1
№10	63.3	0.5	10.4	7.1	2.9	·	3.6	12.2
ЛГ-2	75.0	3.3	7.9	1.7	·	4.6	0.5	6.8
№23	69.6	2.8	4.0	6.9	·		7.7	9.0
№42	70.6	·	..	·		13.2	16.2	—
AC-1	72.1	—	1.0	5.8	3.8		1.2	16.1

Relationships similiar in form to these equations satisfactorily describe the temperature dependence of the electrical conductivity of glasses [9, 10]. Equations (9) and (10) are just as accurate as the above noted viscosity equations and are distinguished by their ease of solution. All calculations according to the indicated equations on the basis of the experimental data were accomplished using a "Razdan-2" computer at the Computer Centers of the Armenian SSR Academy of Sciences and Yerevan State University.

Figures 1 and 2 depict the lg κ — lg η graphs for several of the tested glasses over a wide temperature range encompassing viscosities of 10^2 to 10^{14} poise. Roman numerals on the figures

indicate the zero point on the lg κ axis. We can conclude from the given graphs: 1. The function lg κ − lg η generally is not linear for the indicated interval of viscosities, that is, relationship (1) is not observed. 2. Relationship (1) is valid in the high temperature range (the molten state) which is in agreement with literature data. 3. Relationship (1) also is observed in the anomalous temperature range (viscosity 10^8–10^{14} poise) but with different values for n and C.

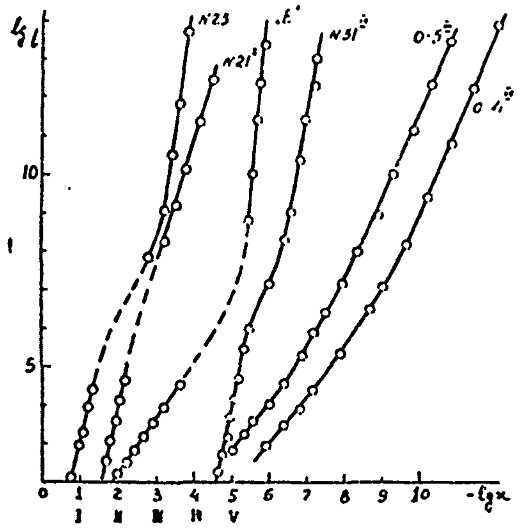

Fig. 1.

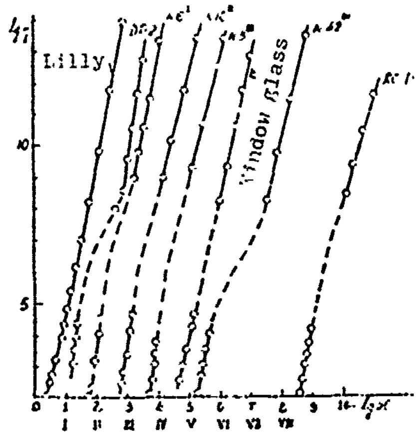

Fig. 2.

Table 2 gives the values for n and C of relationship (1) for the anomalous temperature range and molten-state temperatures of glass.

It is clear from the table that in both cases n > 1 and that on the average the values of n for the anomalous temperature range differ only slightly from n for the molten state. No hard and fast rules are observed in the variation of n, either in the dependence of transition from the molten to the highly-viscous state, or in the composition of the glass.

Table 2. Coefficients of equation $\lg \eta + n \lg \kappa = C$.

Glass	Anomalous temperature range (10^8-10^{14} poise)			Molten State (10^2-10^5 poise)		
	n	C	Δср%	n	C	Δср%
•E..	8.083	--34,94	0,03	1,529	-0.94	2.61
№23	4.610	--5.41	2,64	4.131	-1.02	1.33
№42	4,073	--6.38	0,07	5.316	-7.00	1.28
ДГ-2	5,081	--5,68	2,66	7.115	--5.49	1.04
№6	4,854	-2,05	0,35	6.770	3.16	2.29
№10	3,976	-0,04	0,28	5.706	--2.21	3.51
№3	3,831	0,91	0,51	6.395	--2.32	1.76
№31	5.314	--4,65	1.25	4.471	-0.61	1.41
Лилли *	5,041	1,06	0,90	3.882	-0.70	0.76
№21	3,262	0,76	1,39	4.391	-0.43	0.65
Оконное *	4.333	--0,67	1,02	4.110	-0.22	1.02
АС-1	3,002	2,12	0,79	4,578	-0.68	1.66

Lilly *
Window **

It is clear from the given results that the changes in activation energy of viscosity and electrical conductivity of glasses with transition to the anomalous temperature range is generally irregular. This shows that the processes of viscous flow and the mobility of electrically-conducting cations occur by various and not mutually independent mechanisms. The fulfillment of relationship (1) in the annealing temperature range is also a formal corollary of the fact that the temperature dependence of viscosity and electric conductivity in the indicated temperature range is subject to exponential relationships. Indeed, as the investigation of the temperature dependence of viscosity and electrical conductivity in the annealing temperature range has whown, this dependence is also subject to exponential relationships (2) in which values A_η, A_κ, E_η, E_κ have different values than those for the molten state [5, 6, 9].

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